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Personal exposure to ultrafine particles: the influence of time-activity patterns

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Abstract

Exposure to ultrafine particles (UFPs) is deemed to be a major risk affecting human health. Therefore, airborne particle studies were performed in the recent years to evaluate the most critical micro-environments, as well as identifying the main UFP sources.

Nonetheless, in order to properly evaluate the UFP exposure, personal monitoring is required as the only way to relate particle exposure levels to the activities performed and micro-environments visited.

To this purpose, in the present work, the results of experimental analysis aimed at showing the effect of the time-activity patterns on UFP personal exposure are reported. In particular, 24 non-smoking couples (12 during winter and summer time, respectively), comprised of a man who worked full-time and a woman who was a homemaker, were analyzed using personal particle counter and GPS monitors. Each couple was investigated for a 48-h period, during which they also filled out a diary reporting the daily activities performed. Time activity patterns, particle number concentration exposure and the related dose received by the participants, in terms of particle alveolar-deposited surface area, were measured.

The average exposure to particle number concentration was higher for women during both summer and winter (Summer: women 1.8×10^4 part. cm^{-3}; men 9.2×10^3 part. cm^{-3}; Winter: women 2.9×10^4 part. cm^{-3}; men 1.3×10^4 part. cm^{-3}), which was likely due to the time spent undertaking cooking activities. Staying indoors after cooking also led to higher alveolar-deposited surface area dose for both women and men during the winter time (9.12×10^2 and 6.33×10^2 mm^2, respectively), when indoor ventilation was greatly reduced. The effect of cooking activities was also detected in terms of women's dose intensity (dose per unit time), being 8.6 and 6.6 in winter and summer, respectively. On the contrary, the highest dose intensity activity for men was time spent using transportation (2.8 in both winter and summer).

Keywords

Alveolar-deposited surface area, ultrafine particles, indoor environment, cooking-generated particles, particle dose.

1. Introduction

Airborne particles are related to a range of adverse health outcomes on the human cardiovascular and respiratory systems (Cesaroni et al., 2013; Kreyling, 2006; Pope III and Dockery, 2006; Schmid et al., 2009). The potential of particles to generate adverse respiratory and systemic health effects is related to their capacity to enter the lungs, potentially carrying several toxic compounds with them. At present, it is not known which particle size, morphology or chemical component is most strongly related to the adverse outcomes on human health and further research in this field is required. In terms of particle size, attention has shifted from mass (PM_{10} or $PM_{2.5}$, mass concentration of particles smaller than 10 µm and 2.5 µm in aerodynamic diameter, respectively, collected on a filter) to surface area and particle number concentrations

(Cauda et al., 2012; Franck et al., 2011; Giechaskiel et al., 2009), whose prevalent contribution is from ultrafine particles (UFPs), namely particles with a diameter less than 100 nm. Recent interest in UFPs is due to their high deposition fraction (International Commission on Radiological Protection, 1994), large available surface area, potential to translocate to the circulatory system (Weichenthal, 2012) and ability to induce inflammation. The main challenge for the scientific community working in the fields of air quality and epidemiology is to provide an adequate evaluation of the UFP dose-response relationship (Sayes et al., 2007), which is no easy task, since it requires the accurate measurement of personal exposure levels to UFPs.

The most common approach (applied to PM_{10} and $PM_{2.5}$ monitoring from a regulatory point of view) assumes that each person in a given region has the same exposure level, which is often obtained from a few air quality monitors and reflects the mean concentrations in the entire urban area or community (Buonanno et al., 2010; European Parliament and Council of the European Union, 2008). This approach could lead to significant errors in estimating the exposure of an individual to air pollutants, because actual exposure is strongly related to the time-activity patterns of an individual (Buonanno et al., 2011; Buonanno et al., 2012), followed by their distance from each particle source (Buonanno et al., 2009; Kaur et al., 2005a; Kaur et al., 2005b). In fact, several authors have shown that short-term fluctuations in aerosol concentrations increase morbidity and mortality (Brugge et al., 2007; Strak et al., 2010) and therefore, averaging the values of air pollutant concentrations, which can actually hide peak values, may result in unreliable estimates of exposure (Manigrasso and Avino, 2012; Manigrasso et al., 2013). These are fundamental problems which can only be overcome through personal sampling which is able to monitor the particle concentrations to which people are exposed in every micro-environment they visit during a typical day, together with the investigation of people by age, gender, socioeconomic status, activity level or ethnicity. In fact, personal exposure studies provide a detailed foundation for larger scale exposure and public health studies and this level of detail is substantially different from current methods to generate population level exposure estimates based on fixed-site monitoring networks (Steinle et al., 2013). Several studies have already analyzed the relationship between personal exposures and concentrations measured at fixed monitoring stations, showing substantial differences (Avery et al., 2010; Gulliver and Briggs, 2004). Differences between people can be explained by the time activity pattern of the individuals, as well as the environments in which they spend their time. In fact, even people living in the same location can experience different exposure profiles and short-term exposures, which may contribute significantly to daily average exposure.

The aim of the present work was to characterize a couple's daily exposure to UFPs when living in the same house. High temporal resolution data were linked to detailed time activity patterns in order to evaluate the impact of activity patterns on personal exposure. For this purpose, the couples comprised of a man who worked full-time and a woman who was a homemaker, in order to give two groups of people (man and women) with highly different time activity patterns.

2. Materials and methods

2.1 Study design

The measurements were carried out on weekdays in Central Italy (Southern Lazio, in the macro-area of Frosinone) during summer and winter in 2012. A total of 48 participants (24 couples comprising a man who was working full-time and a woman who was a homemaker) were asked to carry a device to measure particle number concentrations and a GPS to record the micro-environments in which they spent their time. In total, 12 of the couples participated in measurements during the summer and winter, respectively, and all of the full-time workers and homemakers were male and female, respectively. We chose couples where the man worked full-time in order to capture certain activities/micro-environments, including transportation, working in an urban office environments and, more generally, environments not encountered by their female partners. The authors point out that the groups were identified by the time activity patterns (home and full-time workers) and not by the gender.

All participants performed their regular activities and the monitored days were representative of their usual weekdays. Furthermore, they were requested to complete a diary in order to record the activities they carried out throughout the day. Only non-smoking couples were included, 10 of which lived in an urban area, eight lived in a suburban zone and six couples lived in a more rural area. A summary of the full-time worker occupations and the couples' house location is provided in Table 1.

2.2 Instrumentation and quality assurance

The mobile experimental apparatus was composed of two hand-held UFP counters (NanoTracer, Philips) equipped with GPS tracking. This device is based on diffusion charging and it is able to measure the number particle concentration in the 10-300 nm size range by means of the current induced by previously charged particles collected on a filter inside a Faraday cage. The NanoTracer can also estimate the different fractions of lung deposited surface area through a semi-empiric algorithm implemented by Marra et al. (2010). The instruments were used in "advance mode", where particle number concentration measurements were performed every 16 s. These personal monitors are equipped with an internal rechargeable lithium-ion battery, which allows them to be used during outdoor trips.

The counters were calibrated at the beginning of the experimental campaign, in order to allow for data quality assurance, by comparison with: i) a Condensation Particle Counter (CPC, TSI Model 3775) to measure particle number concentration; ii) a Nanoparticle Surface Area Monitor (NSAM, TSI Model 3550) to assess the human lung-deposited surface area of particles (reported as $\mu m^2\ cm^{-3}$) corresponding to tracheobronchial (TB) and alveolar (A) regions of the lung; and iii) a Scanning Mobility Particle Sizer (SMPS, TSI Model 3936) spectrometer to measure the mean diameter of the particle number size distributions. The calibration was carried out within a closed volume space (about 16 L), with uniform and stationary particle number concentration. Details are reported in Buonanno et al. (Buonanno et al., 2013b). Quality assurance of the CPC measurements was guaranteed through calibration and flow checks conducted at the start of the monitoring periods. Each CPC was calibrated in the European Accredited Laboratory at the University of Cassino and Southern Lazio by comparison with a TSI 3068B Aerosol Electrometer (Stabile et al., 2013b).

2.3 Methodology description

Each person kept the NanoTracer device on a belt (with the exception of sleeping time when it was placed in the bedroom) for two days, carrying it with them in all of the micro-environments where he or she spent their time. The subjects were also asked to record their main indoor and outdoor activities, indicating the start and end times for each activity. An analysis of the GPS data in conjunction with the diaries allowed to estimate in an exact way the time spent in each microenvironment, in particular in transportation.

Based on the time duration of each activity, the corresponding average particle number concentration, diameter, and deposited alveolar and tracheobronchial surface area concentrations were calculated. The dose (in terms of deposited alveolar or tracheobronchial surface area) received by subjects in each micro-environment/activity was determined by multiplying the alveolar and tracheobronchial surface area ($S_{a,tb}$) for the time spent (t) in the j^{th} micro-environment and the inhalation rate ($IR_{activity}$) corresponding to the activity carried out (Klepeis, 2006). Then the partial doses were added to estimate the daily total deposited alveolar and tracheobronchial surface area (dose), $\overline{S_{a,tb}}$, as reported in eq. (1).

$$\overline{S_{a,tb}} = \sum_{j=1}^{n} \left(IR_{activity} \cdot S_{a,tb} \cdot t \right) \qquad (1)$$

Inhalation rates for the different activities based on the US EPA approach (U.S. Environmental Protection Agency, 2004).

In order to compare the exposure (dose) in different micro-environments, we determined the “exposure (dose) intensity” linking the daily exposure fraction with the daily time fraction, as described in eq. (2) (Wang et al., 2011) and applied in our previous papers (Buonanno et al., 2011; Buonanno et al., 2012):

Exposure (dose) intensity = Daily exposure (dose) fraction (%) / Daily time fraction (%) (2)

3. Results and discussion

3.1. Study population and daily time activity analysis

The age of all participants was between 25 and 60, 50% of which were men and women, respectively. Figure 1 shows the time spent by participants undertaking each activity.

The average value for total time spent at home was about 1033 min (72%), with women (homemakers) spending more time at home (1211 min, 84%) than their male (working full-time) counterparts (855 min, 59%). The total time spent using transportation was reasonably constant and equal to about 64 min (4%) and 96 min (7%) for homemakers and full-time workers, respectively. The obtained values were in good agreement with those reported by previous studies. For example, Chau et al. (2002) showed that individuals from Hong Kong (China) spent an average of 86% of their time indoors and 3-7% in enclosed transit. Brasche and Bischof (2005) carried out an analysis of the time spent indoors at home in Germany. The mean time spent at home was equal to 942 min (65%) with higher values for women (996 min, 69%) than men (882 min, 61%). This difference was very pronounced in the age range 25-54 years. The overall mean time spent at home was also in good agreement with results from American (940 min, 65%) and Canadian (950 min, 66%) human activity surveys carried out in the 1990's, as reported by Leech et al. (2002). Dons et al. (2012) carried out a personal monitoring study to evaluate differences in Black Carbon (BC) exposure for full-time workers and homemakers, reporting that each group spent 8% (112 min) and 6% of their time using transportation, respectively. Therefore, a comparison between the presented data and the current literature indicates that the Italian lifestyle is typical of that in other Western countries.

3.2. Personal exposure to ultrafine particles

Table 2 presents the average daily personal exposure to ultrafine particles for all 48 participants over a 2-day period, for both summer and winter time. The measured summer exposure values were lower than those for the winter time, and as reported by Buonanno et al. (2013a) and Stabile et al. (2013a), the seasonal variability of airborne particle number concentration was affected by the temperature inversion phenomena. In particular, low incoming solar radiation in the winter time often resulted in aerosol concentrations that peaked at ground level during morning rush hours and remained relatively high throughout the day, due to inefficient ventilation. Furthermore, reduced air exchange rates during the winter time increased the contribution of indoor sources, such as cooking activities (Buonanno et al., 2011). The daily values reported in this work highlight the differences in exposure between members of the same household, during both summer and winter time. The difference between the personal exposures of a full-time worker versus a homemaker of the same household amounts to more than 50%, with the homeworker being more exposed with respect to the full-time worker. These differences were tested using the ANOVA tool and when a significant difference was observed, intergroup comparisons were made using the Student's t test. The analysis of the average daily personal exposure to UFPs exhibited a p-value of <0.05: this can be considered significant for the intergroup comparison analysis. Therefore, homeworkers and full-time workers can be treated as different population groups as their different activity patterns entail strongly different exposures/doses to particles. The typical daily pattern of a couple (Couple 6), living in a rural area during winter, is shown in Figure 2. The first peak for the couple is seen during breakfast time, followed by high particle concentrations during commuting for the full-time worker. During the afternoon and evening, two major peaks are clearly shown (mainly for the homemaker) and these correspond to cooking activities during lunch and dinner time. In fact, cooking activities were observed to affect UFP levels during both eating time, as well as during other periods of time spent at home.

3.3. Dose results

In Table 3 average alveolar deposited surface area dose experienced by the 48 participants is reported for both summer and winter time. The daily alveolar deposited surface area dose of homemakers (women) was equal to $5.40\times10^2 \pm 3.64\times10^2$ mm^2 and $9.12\times10^2 \pm 4.56\times10^2$ mm^2, for summer and winter time, respectively. The daily dose was considerably higher in winter, due to both a greater outdoor particle concentration (resulting from the temperature inversion phenomena) and higher contributions from indoor sources (resulting from low air exchange rates in houses using natural ventilation). The corresponding values for full-time workers (men) were equal to $5.73\times10^2 \pm 2.16\times10^2$ mm^2 and $6.33\times10^2 \pm 3.72\times10^2$ $mm^{2,}$ respectively. A slight increasing trend from summer to winter time was also observed for full-time workers, where the summer values were comparable to those for homemakers (women) and the winter values were much lower.

Overall, the daily doses were in good agreement with the ones obtained by Buonanno et al. (2011) and Buonanno et al. (2012), where activity patterns were combined with micro-environmental data using the Monte Carlo method, and the alveolar-deposited surface area received by different age groups in Italy was estimated, with mean values of 4.1×10^2 mm^2 and 3.8×10^2 mm^2 found for adult women and men, respectively.

Table 3 also reports the dose intensity and contribution to daily dose of the different activities/micro-environments, in terms of alveolar deposited surface area. With regard to the daily dose fractions, it was found that time spent indoors was the major contributor both for homemakers and full-time workers, ranging from 89-97%. An important contribution for homemakers arose from cooking time (27%-28%), corresponding to more than 200 mm^2 during the winter time. This activity also presented the highest dose intensity (equal to 6.6 and 8.6 in summer and winter, respectively), highlighting the very high dose received per time unit during cooking time. The contribution of cooking activities was not limited to cooking time itself, since homemakers were also exposed to high concentrations during eating and other time spent at home. This is because cooking-generated particles in indoor environments are likely to remain airborne long after cooking activities have ceased, in particular when sufficient air exchange ratios are not guaranteed. Another important contribution arose from sleeping time, which, despite making similar daily contributions to dose, was totally different in terms of exposure. In fact, sleeping was characterized by low particle concentrations over a long duration, while cooking was characterized by high concentrations over a shorter time period.

At this point, the authors wish to note that absolute dose could be a misleading parameter when comparing groups with different air-tissue interfaces (alveolar surface area of the lung exposed to air) are considered. Therefore, a lower absolute particle dose in women could still lead to a higher deposited particle "density" in the alveolar region. To this purpose, the average daily doses, normalized according to the air-tissue interface data obtained from Dunnill (1962) and Thurlbeck (1982) are also reported in Table 3. The data clearly show that whatever gender (male *vs*. female) was considered, the normalized deposition (based on the actual available air-tissue interface for women) was always higher than for men, in particular during the winter time.

4. Conclusions

This work evaluated the influence of time-activity patterns on the personal exposure of 24 Italian couples (12 in winter and 12 in summer) to UFPs based on their time activity patterns, in terms of time spent in each micro-environment over a two day period. Non-smoking couples, where the man worked full-time and the woman was a homemaker, were chosen to undertake the experimental analysis. Each couple carried a GPS and a personal diffusion charger monitor to measure particle number concentrations, as well as alveolar-deposited surface area concentrations for each participant. Alveolar-deposited surface area dose was also calculated on the basis of the activities performed.

Time activity pattern data indicated a higher mean time spent at home by women compared to men (996 min *vs*. 882 min). The average particle number concentrations experienced by women were also higher (roughly twice) than men, both during summer (1.8×10^4 *vs*. 9.2×10^3 part. cm^{-3}) and winter time (2.9×10^4 *vs*. 1.3×10^4 part. cm^{-3}). Average values were higher in winter due to both reduced indoor ventilation and the frequent occurrence of outdoor inversion phenomena.

With regard to particle alveolar-deposited surface area dose, higher values were measured for women than men during the winter time (9.12×10^2 *vs.* 6.33×10^2 mm^2), whereas summer values were quite similar for both men and women. The higher dose received by women was mainly due to cooking activities, which also resulted in high concentrations in indoor environments long after the cooking activity had ceased. This clearly explains the reason why in winter (reduced ventilation), the difference in dose between men and women was larger. In fact, the dose intensity (dose per unit time) of cooking activities was the largest amongst all of the activities performed (8.6 and 6.6 for women in winter and summer time, respectively). The highest dose intensity activity for men was during the use of transportation (2.8 both in winter and summer).

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Tables

Table 1 - Population group characteristics in terms of full-time worker occupation and couples' house location.

Summer time			Winter time		
Couple	Full-time worker	House location	Couple	Full-time worker	House location
1	factory worker	urban	13	employee	suburban
2	employee	urban	14	employee	rural
3	salesman	suburban	15	employee	urban
4	employee	urban	16	salesman	suburban
5	odontology	rural	17	factory worker	urban
6	odontology	rural	18	laboratory technician	rural
7	employee	urban	19	employee	suburban
8	policeman	suburban	20	medical practitioner	urban
9	gym trainer	rural	21	engineer	urban
10	salesman	urban	22	lawyer	urban
11	barman	suburban	23	professor	suburban
12	employee	suburban	24	employee	rural

Table 2 - Average daily personal exposure to ultrafine particles (part. cm-3) of all subjects during summer and winter time. A p-value < 0.05 was considered significant.

season	**Homeworker women**	**Full time worker men**
Summer	$1.8\times10^4 \pm 1.0\times10^4$	$9.2\times10^3 \pm 3.0\times10^3$
Winter	$2.9\times10^4 \pm 7.9\times10^3$	$1.3\times10^4 \pm 1.0\times10^3$

Table 3 - Dose, dose intensity and contribution to the daily dose of the different activities/micro-environments in terms of alveolar deposited surface area for homeworkers (women, W) and full time workers (men, M).

Micro-environment or activity	**Daily dose fraction (%)**				**Dose Intensity**			
	Summer time		Winter time		Summer time		Winter time	
	W	M	W	M	W	M	W	M
Indoor	41	23	44	22	0.9	1.0	1.0	1.1
Transportation	3	9	2	8	0.8	2.8	0.7	2.8
Eating	6	5	8	6	1.8	1.6	2.3	1.6
Cooking	27	1	28	1	6.6	0.8	8.6	1.2
Sleeping	19	20	17	21	0.5	0.7	0.6	0.7
Working	0	41	0	39	0.0	0.9	0.0	0.8
Outdoor	4	1	1	3	0.3	0.1	0.2	0.2
Daily alveolar deposited surface area dose (mm^2)	540±364	573±216	912±456	633±372				
Normalized daily alveolar deposited surface area dose ($mm^2\ m^{-2}$)	9.2	8.2	15.5	9.1				

Figure captions

Figure 1 - Statistics of the time spent by homeworkers (women, W) and full-time workers (men, M) on each activity: box-plots report median, 1st (Q_1) and 3rd (Q_3) quartile, minimum and maximum values. Upper (U) and lower (L) whiskers were evaluated as $U=Q_3+1.5\times(Q_3-Q_1)$ and $L=Q_1-1.5\times(Q_3-Q_1)$, respectively. Measurement data higher than the “upper whisker” or lower than the “lower whisker” were considered outliers, they are not showed here.

Figure 2 - 24-h particle number concentration trends of couple 6 (living in a rural area) during winter time.